NEUTRON YIELDS FROM PROTON-INDUCED

SPALLATION REACTIONS IN THICK TARGETS OF LEAD

M. A. Lone and P. Y. Wong AECL Research, Chalk River Laboratories Chatk River, Ontario, Canada KOJ IJO Phone (613) 584-3311

ABSTRACT

Total neutron yield, integrated over the neutron energy and emission angle, is calculated for spallation reactions in lead at proton energies between 0.4 and 2 GeV. The computations were made with the LAHET/MCNP code package using the intranuclear-cascade-evaporation model of Bertini. The dependence of the integrat neutron yield on the target dimensions is investigated and the contribution from the (n,xn) interactions of neutrons of energy < 100 MeV is evaluated. For 1.6 GeV protons, the integral neutron yield from a 100 cm diameter, 100 cm long Pb target is calculated to be in the range of 46 to 57 n/p, depending on the level density parameter used in the evaporation model.

I. INTRODUCTION

Evaluation of the integral neutron yield from spallation reactions has been the subject of many studies, ⁷¹⁻¹ The Intranuclear-Cascade-Evaporation (INCE) model of Bertini⁴ has proved to be reasonably successful in describing the spallation reaction process. The integral neutron yield computed with this model for spallation reactions in Pb targets at proton energies between 0.5 and 1.5 GeV is found to be within 20% of the measured yield. However, the calculated yield using traditional parameters for level density was found to be consistently higher³, 14 than the measured yields.

In the INCE model of Bertini, the elastic collisions of nucleons and pious with nuclei are ignored. The computation of the nonelastic collision is based upon the premise that at energies above a few tens of MeV, the interaction of a nucleon or pion incident upon a complex nucleus can be treated as the interaction of the incident nucleon/pion with the individual nucleons of the nucleus. This premise holds because the de Broglie wavelength of the incident particle is comparable to or smaller than the average distance between the nucleons within the nucleus.

A nonelastic collision is modeled to take place in two successive stages. In the first stage-- the intranuclear cascade-the interactions with the individual target nucleon create collision products of high-energy particles. These may interact with other single nucleons inside the same nucleus (intranuclear cascade) or inside other nuclei (internuclear cascade), In the second stage--the evaporation process--the excited residual nucleus remaining at the end of the intranuclear cascade is assumed to have its residuat excitation energy equipartitioned among the remaining nucleons, and the nucleus cools off by evaporation of low-energy, low-mass particles until the excitation energy is <7 MeV,when the evaporation of particles is suppressed by binding energy. The remaining energy is emitted as gamma rays. The INCE model incorporated in the LAHET/HETC family of Monte Carlo codes includes the possibility of fission during the second stage, in competition with the evaporation process. Details of the nuclear model and the associated parameters used in the LAHET/HETC version of the INCE model are discussed in several publications .4- 14

For projectile energies above 100 MeV, a comparison¹⁵ of the INCE model predictions of the differential neutron yield with measured data shows that the model predicts relatively correct neutron production in the evaporation region, but underpredicts neutrons of energy above 50 MeV, especially at large scattering angles. At lower projectile energies discrepancies are progressively larger, as exemplified by the comparison of neutron spectra] ⁶ from ²⁰⁸Pb(p,xn) reactions at 15 MeV proton energy.

The concern over the energy range of validity of the INCE model of Bertini focuses on its neglect of the nuclear structure effects in the nonelastic interactions. It is generally assumed that the code modeling of interactions using the INCE model at projectile energies much below 100 MeV is questionable. This energy regime is important when considering the transport of nucleons in thick targets, in which a given nucleon may suffer several collisions and concomitant kinetic energy degradation, In addition, secondary nucleons of energy below 100 MeV are generated by the nonelastic interactions at higher energies, Atthough the prediction of differential yields shows an increasing discrepancy at lower projectile energies, this may not necessarily apply to the prediction of the integral neutron yield, since many reaction channels contribute and deficiencies in the differential yield may cancel out. The Bertinimodel has been used for energies down to 15 MeV with reasonable success in predicting the iutegrat neutron yield to within 20% of the empirical yields. 3,14

In this paper, we present results from calculations of the integral neutron yields from spallation reactions at proton energies between 0.4 and 2 GeV in cylindrical Pb targets of diameters ranging from 10 to 200 cm. The leakage of nucleons from targets is evaluated and the overall contribution of neutron interactions at energies below 100 MeV is estimated. The sensitivity of the computed yields to the level density parameter is examined and the predictions are compared with the empirical data.

II. COMPUTATION DETAILS

Computations were made with the LANL code package ¹⁰ LAHET/MCNP using the traditional INCE model of Bertini,⁴ supplemented by the RAL model⁹ to describe the fission induced by high-energy interactions. Transport of the particles included the multiple Coulomb scattering of primary protons and the elastic scattering of neutrons with energies below 50 MeV. The cascade histories were computed through all generations. The dependence of the integral neutron yield on the level density parameters was explored.

The computational geometry considered a cylindrical Pb target with an on-axis pencil beam of incident protons or neutrons. The calculations were made for various diameters and lengths of a Pb target, The LAHET code followed all interactions of protons, pions, and muons. Neutrons were followed in LAHET down to a cutoff energy of 20 MeV and were passed on to the MCNP code as source particles for transport in the target.

The total number of neutrons of all energies escaping from all surfaces of the bare target following a combined LAHET/MCNP treatment were tallied and defined as the integrated neutron yield available from the spallation target as a neutron source. This tall y does not inchrde the neutrons that were captured in the Pb target, The spectral energy distributions of the neutrons and protons escaping from the target were also tallied.

For a benchmark comparison with the measured data, the set-up of the experiments ¹⁷ was modeled and the neutrons captured in the H20 moderator surrounding the Pb spallation targets were tallied. Potential sources of uncertainty in estimates of the total yield were investigated. These included the leakage of neutrons from the moderator assembly, the fraction of neutron yield captured in the region of the moderator in which the neutron flux was actually measured, and the effect of the beam size on the leakage of protons and high-energy neutrons from the Pb target.

III. RESULTS

A. Integrated Neutron Yield and (n,xn) Multiplication

Figure 1 shows the calculated integrated yield of neutrons escaping from a cylindrical Pb target of 100 cm diameter and 100 cm length after bombardment with neutrons and protons. The solid curve represents a neutron and the dashed curve a protoo projectile. These computations were made using the

RAL fission-evaporation model $^{7-10}$ and the default level density option in the code '0 (see section III D). The data points show the measured yield from proton-induced reactions reported in the literature. 9 ~20



Figure 1. Total neutron yields calculated for proton and neutron projectiles on Pb targets. The data points are: solid circle at 32 MeV from Ref. 19, open circle at 100 MeV from Ref. 20, and stars between 0.5 to 1.5 Ge V from Ref. 18. The latter are for a 20 cm diameter, 60 crn long target.

For neutrons of energy below 20 MeV, the yield is computed with the MCNP code using the evahrated cross sections. In this energy range, the neutron multiplication is produced by the (n,2n) reaction that dominates (86% at 15 MeV) the total nonelastic cross section, including all of the $(n,n'\gamma)$ and the (n,γ) channels, A 15 MeV neutron incident on an infinite block of Pb can generate neutrons until its energy drops to the neutron binding energy of $\approx 7 \text{ MeV}$. Each neutron generated would account for an energy loss of 8 MeV, if we adopt 1 MeV average energy for the outgoing evaporated neutron. Thus all interactions initiated by a 15 MeV incident neutron would generate ((15-7)x0.86/8) ≈ 0.9 additional neutrons inside a large Pb target.

For energies above 20 MeV, the calculations are made with the INCE model, and the rise in the solid curve just above 20 MeV reflects the onset of (n,xn) reactions with x>2. Table 1 lists the distribution of the residual nuclei produced by all interactions of a 30 MeV incident neutron. It is clear that many channels contribute to the neutron production. The cross sections of these reaction channels are not established fully in this energy range, and the magnitude of the rise in the neutron yield curve needs to be validated with integral experiments.

For protons, the data shown by the dashed curve (Figure 1) is calculated by the INCE model. In this case, the lower yield is due to the loss of kinetic energy of protons by the ionizing atomic collisions that reduces their range in the target.

The data point at 32 MeV shows the measured total yield of 0.16 n/p reported by Tai et al.²⁰ and at 100 MeV the yield of 0.34 n/p measured by Lone et al.²¹ The degree of agreement clearly indicates that the model is reasonably successful in predicting the totaf neutron yield, in spite of any deficiency in predicted differential yields. The data points between 0.54 and 1.5 GeV show the measured¹⁷ yield from a Pb target of 20 cm diameter and 60 cm length, These data are discussed in section 4.

Table 1								
Neutron-producing	reaction	channels	induced	in	208 Pb	by	a	30
May neutron projectile								

Channel	Residual	Yield
(n, n')	8 Pb	2.0E-1
(n, 2n)	207Pb	1.2E-1
(n, 3n)	206Pb	3.1E-1
(n, 4n)	205Pb	5.3E-1
(n, pn)	207_{Tl}	2.3E-2
(n, p2n)	206TI	2.7E-2
(n, p3n)	205TI	4.2E-3



Figure 2. The variation of the calculated total neutron yield with the target diameter.

B. Leakage of High-Energy Nucleons from Finite-Size Pb Targets

Figure 2 shows the variation in the integrated neutron yield with the target diameter. The yield increases steadily and reaches a plateau at a diameter of about 100 cm. This change is due to the propagation and interactions of the high-energy neutrons that escape from smaller-diameter targets, as was pointed out earlier by Bowman et $al.^2$ We checked this by recording the spectra of the high-energy nucleons leaking from

the targets and computing their potential contributions with the data from Figure 1.

Figure 3 shows the total number of protons escaping from the targets. The sharp decrease with increasing target diameter is due to the suppression of the leakage from the circumference. The residual at diameters above 50 cm is due to the leakage from



Figure 3. The total proton leakage as a function of the target diameter,



Figure 4 The cumulative energy spectrum of protons escaping from the target.

the front and back ends of the target. Examples of the cumulative spectral distributions (total number of protons above energy E) of protons leaking from the targets are shown in Figure 4. When a 1.6 GeV proton is incident on a 60 cm long

Pb target, the leakage from targets of 10, 20 and 100 cm diameters is, respectively, 0.28, 0.13, and 0.076 protons per projectile. The energy of protons escaping from the 10 cm diameter target extends almost to the incident energy, whereas the energy of protons escaping from the 20 and 100 cm targets shows a minimum energy loss of about 0.7 GeV. The majority of these escape from the back end without undergoing a nonelastic nuclear collision. The loss of neutron yield due to the leakage of these protons is calculated to be 3.4, 1.8 and 1.4 Or/projectile), respectively, for targets of 10, 20 and 100 cm diameters.

Figure 5 shows the leakage of high-energy neutrons as a function of the target diameter. As expected, this is significantly higher than the leakage of protons. When a 1.6 GeV proton is incident on a 60 cm long Pb target, the number of neutrons of energy >20 MeV that escape from targets of diameters of 10, 20 and 100 cm is, respectively, 2.8, 2.3 and 0.8, Figure 6 shows that the majority of these neutrons have energies below 100 MeV. The reduction in the integrated neutron yield due to the escape of these neutrons is calculated to be 23, 18.3 and 6.5, respective y, for the 10, 20 and 100 cm diameter targets.



Figure 5. The total leakage of neutrons of energy above 20 MeV.

C. Neutron Gain from Interactions of Neutrons of Energy Below 100 MeV

The above analysis emphasizes the contribution from neutron interactions at energies below 100. For a 1.6 GeV proton projectile on a Pb target, Figure 7 shows the net contribution to the totat neutron yield from neutrons of energy up to 100 MeV. All neutrons of energy less than 100 MeV generate 15.3 neutrons in a 100 long, 100 cm diameter target. The equivalent contribution to a 60 cm long, 20 cm diameter target is 5.6 neutrons.



Figure 6. The cumulative energy spectrum of neutron escaping from the target.



Figure 7. The contribution to total neutron yield from interactions of neutrons of energy below En.

D. Level Density Dependence

In the evaporation model^{7,22} the probability of emission of a particle i of kinetic energy E is expressed as

Pi(E) \propto g₁ml E σ_{ic} (E) ω (E*)

where g_i is the number of spin states, ml is the mass of the emitted particle and σ_{iC} (E) is the cross section for the compound nucleus formation in the inverse reaction. The level density for excitation energy E* is given by

$$\omega(E^*) = \omega_{a}e^{(2(aE^* - \delta)^{1/2})}$$

where ω_0 and a are constants for a given nucleus and δ is the pairing energy. The parameter a depends on the mass number A. The evaporation model incorporated in the HETC code uses the value a $\approx A/B_{o}$, where B. is an input parameter, various reviews of the data on level densities indicate B. to be in the range of 8 to 20. However, the data show that the level densities are strongly influenced by nuclear shell structure, and the densities of magic nuclei are several orders of magnitude lower than the densities for the mid-shell nuclei. The Julich option in LAHET incorporates the shell-structure-dependent B_0

No nuclear-structure effects are incorporated in the formalism used to calculate the inverse cross section. The $code^{7,22}$ calculates it from the geometric cross section modified by an empirical expression (to take into account energy dependence) and a Coulomb barrier correction for charged particle emission. It has been argued that if the shell effects are included in the level densities, corresponding effects should be included in the inverse cross sections.

The integrated yield of neutrons depends on B_o , and is found to decrease linearly with increasing B_0 . Increasing B. from 8 to 20 decreased the yield by about 24%,

IV. EXPERIMENTAL DATA AND COMPARISON

In the past, the primary interest in **spallation** targets was for designing neutron source facilities for research purposes. Consequently, most of the studies of the **spallation** neutron yield investigated targets of relatively moderate dimensions, to optimize neutron source brightness.

An extensive series of measurements were carried out at the BNL Cosmotron in 1965 by a Chalk River-Oak Ridge team. ¹7 A variety of target materials and shapes, and a range of proton energies (from 0.5 to 1.5 GeV), were used. Six different targets of Be, Sn, Pb and depleted U were studied. The targets were mounted in a large cylindrical tank of H20 (1.83 m diameter by 1.83 m high), which was located in an external beam of the Cosmotron. The absolute neutron flux distribution in the water surrounding the targets was measured, with up to 55 Cu or Au foils mounted on a lucite frame about 110 cm long by 60 cm high. The frame was mounted symmetrically above the target in a vertical plane. The foils were counted by an automatic counting system, which had been calibrated previously against a $47r \beta - \gamma$ coincidence system. As a further check on the system, a standardized neutron source was measured in the Chalk River Pool Test Reactor tank using the same foil array and counting system. The source strength thus determined was within 270 of the value determined by the National Research Council of Canada.

The integrated proton current into the targets was measured by the irradiation of polyethylene foils placed at the entrance of the beam into the water tank. The 11C activity produced in the foils was measured in a calibrated counter at BNL. The neutron yield was evaJuated from the neutron capture rate in the H20 surrounding the target, and making minor corrections for the leakage of neutrons from the tank and parasitic absorption in the target and the structural materials. The overall accuracy of the measured yield is stated to be 5%. However, there are no published details of the neutronic analysis of the experiments. An analysis of the experiment with LAHET/MCNP simulations shows that only 3% of the neutrons would have escaped from the large moderator tank. The fraction captured in the foil region was 9170. Changing the proton beam diameter from 1 cm to 4 cm shows insignificant variation in the calculated leakage of protons or high-energy neutrons from the target. Thus the overall magnitude of the corrections does not appear to be great.

A comparison between the measured data ¹⁷ and the calculated yields is shown in Figure 8 for the 20 diameter, 60 cm long Pb target. The standard deviation of the Monte Carlo computed data is better than 1%. The yield calculated with the mass-dependent B_0 values, shown by the solid curve, is in excellent agreement with the measured data. For nuclei near Pb, the mass dependent value of B. is about 20 and the yield calculated with this value shows similar good agreement, as indicated by the short dashed curve. The yield calculated with the default level density option in LAHET, shown by the medium dashed curve, is about 25% too high. The same is true of the yield shown by the long dashed curve that is calculated for $B_0 = 8$.



Figure 8. Comparison of theoretical and experimental data on total yields from 60 cm long Pb targets of 20 cm diameter. The experimental data is from Ref. 17. The B. values used in the calculations are: solid curve -- mass dependent; short dash -20; medium dash - default in LAHET; long dash --8.

v. SUMMARY

The total neutron yields from thick Pb targets were computed with the codes LAHET/MCNP. The agreement with the empirical data¹⁷ shown in Figure 8 suggests the use of a mass dependent B. parameter. At 1.6 GeV proton energy, the total neutron yield from spallation reactions in a 100 cm long Pb target of 100 cm diameter is calculated to be 57 n/p, for B. = 8 and 46 n/p for B_0 =20.

VI. ACKNOWLEDGMENTS

We appreciate very much the kind help of Richard E. Prael in implementing the LAHET/MCNP code at CRL for these studies.

VII. REFERENCES

1. J.S. FRASER and G.A. BARTHOLOMEW, "Spallation Neutron Sources", in <u>Neutron Sources for Basic Physics and Applications</u>, (cd, S. Cierjacks), Pergaman Press, New York (1983).

2. C.D. BOWMAN et al., "Nuclear Energy Generation and Waste Transmutation Using an Accelerator-Driven Intense Thermal Neutron Source", <u>Nuclear Instruments and Methods in Physics Research. A320,(1992) 336.</u>

3. M.A. LONE, "Thick Target Neutron Yields from Spallation Reactions", <u>AECL Report. AECL-10599 (1992).</u>

4. H.W.BERTINI, "Spallation Reaction: Calculations", Spallation Reactions and Their Applications. eds. B.S.P. Shen and M. Merker. D. Reidel Publishing Company (1976) 27.

5, R.G. ALSMILLER, Jr. "Nucleon-Meson Transport Calculations", <u>Spallation Reactions and Their Applications</u>, eds. <u>B.S.P. Shen and M. Merker, D. Reidel Publishing Company</u> (1976) 139.

6. T. W. ARMSTRONG, "The Intranuclear-Cascade-Evaporation Model", Computation<u>Techniques in Radiation</u>Transport and Dosi metry, eds. W.R. NELSON and T.M. JENKINS. Plenum Press London (1980)3 11,

7. P. CLOTH, D, FILGES, G. STERZENBACH, T.W. ARMSTRONG and D.B.L. COLBORN, "The KFA-Version of the High-Energy Transport Code HETC and the generalized Evacuation Code SIMPEL", KFA Reprt Jul-Spez-196 (1983), ISSN 0343-7639.

8. T, W. ARMSTRONG, P. CLOTH, D.FILGES and R. NEEF, "An Investigation of Fission Models for High-Energy Radiation Transport Calculations", <u>KFA Report Jul-1859(1983).</u>

9. F. ATCHISON, "Spallation and Fission on Heavy Metal Nuclei under Medium Energy Proton Bombardment", in <u>Targets</u> for Neutron Beam Spallation Sources, Jul-Conf-34, Kernforschungsanlage Julich GmbH (1980).

10. R.E. PRAEL and H. LICHTENSTEIN, "User Guide to LCS: The LAHET Code System", Las Afamos National Laboratory Report LA-UR-89-3014 (1989).

11. W.A. COLEMAN and R.G. ALSMILLER, Jr., "Thermal Neutron Flux Generation by High-Energy Protons", <u>Nuclear Science and Engineering 34. (1968) 104.</u>

12. G.J. RUSSELL, J.S. GILMORE> R.E. PRAEL, H. ROBINSONS and S.D. HOWE, "Spallation Target-Moderator-Reflector Studies at the Weapons Neutron Research Facility", Proc_Symposium on Neutron Cross-sections from 10-50 MeV. Brookhaven National Laboratory Report BNL-NCS-51245,V1 (1980) 169.

13. P.M. GARVEY, "Neutron Production by Spallation in Heavy Metal Targets", <u>Proc. of Meeting on Target for Neutron</u> <u>Beam Spallation Neutron Sources.</u> Jul-Conf-34, ISSN0344-5798, Julich (1980).

14. T.W. ARMSTRONG, P. CLOTH, D.FILGES and R.D. NEEF, "Theoretical Target Physics Studies for SNQ Spallation Neutron Source", <u>KFA Report Jul-Spez-120(1981)</u>.

15. W. AMIAN, P. CLOTH, P. DRAGOVITCH> V. DRUKE, D. FILGES and M.M.MEIER, "Neutron Spectra Measurements and Monte Carlo Simulations of (p, xn) Reactions", <u>Nuclear</u> <u>Data for Science and Technology, eds. S.M. OAIM, Springer</u> <u>Verlag (1992) 696.</u>

16. R.G. MILLER, Jr. and O.W. HERMAN, "Calculation of the Neutron Spectra from Proton-Nucleus Novelistic Collisions in the Energy Range 15 to 18 MeV and Comparison with Experiments", <u>Nuclear Science and Engineering 40(1970)254</u>.

17. J.S. FRASER, R.E. GREEN, J.W. HILBORN, J.C.D. MILTON, W.A. GIBSON, R.E. CROSS and A.ZUCKER, Neutron Production in Thick Targets Bombarded by High Energy Protons", <u>Phys. in Canada 21 (1965) 17.</u>

18. V.S. BARASHENKOV, N.M. SOBOLEVSKII and V.D. TONEEV, "The penetration of Beams of High-Energy Particle Through Thick Layers of Materials", <u>Atomnaya Energiva 32</u> (1972)217.

19. J.C.D. MILTON and J.S FRASER, "A Monte Carlo Calculation of Neutron Production in Heavy Element Targets in the Range 0,3 to 1 GeV", <u>AECL Report, AECL-2259 (1965).</u>

20. Y.K. TAI, G.P. MILLBURN, S.K. KAPLAN and B.J. MOYER, "Neutron Yields for Thick Targets Bombarded by 18and 32-MeV Protons", Phys. Rev 109 (1958) 2086.

21. M.A. LONE, R.T. JONES, B.M. TOWNES, L. NIKKINEN and R.B. MOOR, "Total Neutron Yields from 100 MeV Protons on CU, Fe, and Th", <u>Nucl.Instr. and Method. in Physics Research A256 (1987) 135.</u>

22. I. DOSTROSKY, Z. FRAENKEL and L. WINSBERT, "Monte Carlo Calculations of Nuclear Evaporation Processes. IV. Spectra of Neutrons and Charged Particles from Nuclear Reactions", <u>Phys. Rev. 118 (1960) 781.</u>